



Characterizing Sorption and Modeling Phosphorus Movement on Candler and Immokalee Fine Sand

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Abstract

Laboratory and computer experiments were conducted to 1) determine the effect of supporting electrolyte on mass distribution coefficient (K_D) for predicting P movement at 30- and 60-cm soil depths using HYDRUS-1D and 2) compare the temporal P concentrations as a function of soil type. The results on Candler fine sand at Lake Alfred showed that P contents for the K_D estimated with 0.01 M KCl and 0.005 M CaCl₂ were 10 to 15% higher than those predicted with a K_D value measured with fertilizer mixture. The predictions on Immokalee fine sand showed that P contents for the K_D estimated with fertilizer mixture and 0.005 M CaCl₂ were 12 to 20% higher than those predicted with a K_D value measured with 0.01 M KCl. The outputs with K_D measured with 0.005 M CaCl₂ appear to be close to those predicted with a K_D measured with fertilizer mixture on Immokalee fine sand. However, the analysis of the K_D values across all electrolytes on the two soils studied revealed that 0.01 M KCl is the electrolyte that yields K_D values fairly close to fertilizer mixture on Candler fine sand and would be appropriate to use for the coated sand while 0.005 M CaCl₂ was comparable with fertilizer mixture on Immokalee fine sand and would be an appropriate electrolyte to use for such uncoated sands. The results of study will help in improving estimation of K_D s and description of P transport in citrus production systems of Florida sandy soils. This would provide necessary information for sustainable environmental management and reduce problems of eutrophication and prevent groundwater contamination.

Keywords

Phosphorus adsorption, Simulation modeling, Entisol, Spodosol

Introduction

Adsorption is the mechanism that soil cations and anions (including various phosphorus (P) compounds) are retained by soil particles. The adsorption process tends to restrict compound's mobility and bioavailability [1]. Thus, the procedure for determining the P sorption isotherms could then provide information on their mobility in the soil. The supporting electrolyte concentration is chosen to mimic that of soil solution. Most commonly 0.01 M CaCl₂ [2,3], 0.01 M NaCl [4], 0.005 M CaCl₂ [1], 0.1 to 3 mM KCl [5], 0.05 M KCl [6,7], and 0.01 M KCl [8,9] have been used as electrolytes in studies on P and K sorption. Nair, et al. (1984) [10] reported that P sorption varies with ionic strength and cation species of the supporting electrolyte. For example, [10] showed that P adsorption was generally lower with K⁺ as the supporting electrolyte cation compared with Ca²⁺. These studies and others have not explained the rationale behind use of a

particular electrolyte other than equilibrating the solutions in deionized or tap water.

The chemical characteristics of soils dominating the Flatwoods and Ridge regions of Florida are well described in [11] and some were also determined in this study. The soil at the Flatwoods consists of nearly level and poorly drained on the Flatwoods classified as Immokalee fine

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Table 1: Selected soil chemical characteristics for immokalee and candler sand.

Soil	Soil depth (cm)	pH [†]	OM [§]	CEC [‡]	NH ₄ ⁺	NO ₃ ⁻	M1P [†]	M1K [‡]	IN [¶]
Immokalee	0-15	5.6	0.61	6-Feb	3.5	4.9	46.5	15.2	8.4
Immokalee	15-30	5.2	0.41	6-Feb	2.3	4.1	28.7	11.8	6.4
Candler	0-15	5.3	1.96	4-Feb	2.6	8.7	115.8	29.7	11.2
Candler	15-30	4.9	1.56	4-Feb	2.9	5.3	112.8	23	8.2

[†]Soil to water ratio = 1:2 (mass/volume); [§]OM: organic matter expressed as a percentage; [‡]CEC: cation exchange capacity expressed in cmol (+) kg⁻¹ (CEC reported by Obreza and Collins; 2008); [†]Mehlich 1 P (mg kg⁻¹); [‡]Mehlich 1 K (mg kg⁻¹); [¶]IN: Inorganic N (mg kg⁻¹).

Table 2: Soil physical characteristics of the Immokalee and Candler fine sand.

Soil	Depth (cm)	Bulk Density (g cm ⁻³)	[‡] K _{sat} (cm h ⁻¹)	[§] θ _{sat} (cm ³ cm ⁻³)	^{§§} θ _r (cm ³ cm ⁻³)	[†] FC (cm ³ cm ⁻³)	[†] AWC (cm ³ cm ⁻³)
Immokalee	0-15	1.62	15.82	0.343	0.013	0.090	0.077
Immokalee	15-30	1.62	13.97	0.362	0.013	0.100	0.087
Immokalee	30-45	1.59	13.22	0.390	0.013	0.100	0.087
Immokalee	45-60	1.61	14.57	0.318	0.013	0.095	0.082
Candler	0-15	1.65	15.53	0.362	0.009	0.074	0.065
Candler	15-30	1.64	15.94	0.330	0.009	0.104	0.095
Candler	30-45	1.57	14.76	0.313	0.009	0.100	0.091
Candler	45-60	1.68	15.73	0.421	0.009	0.094	0.085

[‡]K_{sat}: Saturated hydraulic conductivity; [§]θ_{sat}: Saturated moisture content; ^{§§}θ_r: Residual moisture content obtained from Obreza, unpublished data; [†]FC: Field capacity moisture content at 10 kPa; [†]AWC: Available water content.

sand (sandy, siliceous, hyperthermic Arenic Haplaquods) with the spodic horizon lying within 1 m from the soil surface. The soil at the Ridge, classified as Candler fine sand (hyperthermic, coated Typic Quartzipsamments) is a well-drained sandy soil with no continuous layer limiting vertical water movement [11]. The Immokalee and Candler fine sand are moderately acidic (pH ranging from 4.9 to 5.6) with > 94% sand textural composition, have low organic matter content (ranging from 0.41 to 0.61% on Immokalee fine sand and from 1.6 to 1.96% on Candler fine sand) and low cation exchange capacity (CEC) (ranging from 2 to 6 cmol (+) kg⁻¹), have inorganic N in the range of 8.2 and 11.2 mg kg⁻¹, moderate to very high P (in the range of 28.7 to 46.5 mg kg⁻¹ for Immokalee sand and 112.8 to 115.8 mg kg⁻¹ for Candler fine sand) and K in the range of 11.8 to 15.2 mg kg⁻¹ for Immokalee fine sand and 23.0 to 29.7 mg kg⁻¹ for Candler fine sand (Table 1) [12]. It has been accepted that soil CEC, particularly the CEC contribution from organic matter content (OMC) has a significant influence on the soil P movement [13]. This study sought to 1) determine the effect of supporting electrolyte on the mass distribution coefficient (K_d) for predicting P movement at 30- and 60-cm soil depths using HYDRUS-1D; and 2) compare the P concentrations with time as a function of soil type.

Materials and Methods

Laboratory adsorption study

The baseline soil chemical and physical properties for soils used in the study are described in (Table 1 and Table 2). Sorption isotherms on the disturbed soil samples (0-15 cm, 15-30 cm) were determined using the batch

equilibration procedure [14]. The initial solution concentrations from potassium dihydrogen phosphate (KH₂PO₄) for P in 0.005 M CaCl₂ were 10, 25, 50 mg PL⁻¹. To determine P sorption isotherms, soil samples were obtained from 5 random positions per site at two depths giving a total of 10 samples. Each sample was weighed in triplicates plus a blank control. A 10 g air-dried, subsample of soil < 2 mm particle size was placed in a centrifuge tube and equilibrated with 20 ml (soil solution ratio 1:2) of 3 initial concentrations of P solutions. The centrifuge tubes were shaken for 24 h, centrifuged for 20 min, and filtered through a filter paper (Whatman, #42). All these procedures were done at room temperature ~25 ± 1 °C as recommended by [14] but the filtrate was later stored at < 4 °C until analyzed for P.

Phosphorus sorption isotherm determination

The solutions from the adsorption study were analyzed for P using ICP-AES and calibration standards of 10, 30 and 50 mg PL⁻¹ preparation in fertilizer mixture (with additions of 0.027 mM NH₄NO₃ and 0.013 mM KCl), 0.01 M KCl and 0.005 M CaCl₂. The amount of chemical sorbed to the soil was calculated from the difference between the initial and equilibrium solution concentration:

$$S_e = \frac{V_o}{m} (C_o - C_e) \quad (1)$$

Where S is the adsorbed concentration (mg kg⁻¹); V_o is the volume of initial solution (L); m is the soil mass (kg); C_o is the initial concentration of the standard solution (mg L⁻¹), and, C_e is the soil solution concentration at equilibrium (mg L⁻¹).

Sorption isotherms for P were calculated using the Freundlich equation:

$$S_e = K_f C_e^N \quad (2)$$

Where K_f is the Freundlich sorption coefficient ($\text{mg}^{1-N} \text{kg}^{-1} \text{L}^N$) and N is an empirical constant related to adsorption phenomena [15]. The linearized form of the Freundlich equation was used to calculate K_f and N :

$$\ln S_e = \ln K_f + N \ln C_e \quad (3)$$

Where S is the adsorbed equilibrium concentration (mg kg^{-1}); C_e is the equilibrium concentration (mg L^{-1}) and C_{\max} is the estimated maximum concentration (mg L^{-1}) and K_f and N are calculated from the intercept and slope of Equation 3. To find average linearized K_D for the Freundlich isotherm, the integrated form of the equation was used:

$$K_D = \frac{\int_0^{C_{\max}} N K_f C^{N-1} dc}{\int_0^{C_{\max}} dc} = K_f C_{\max}^{N-1} \quad (4)$$

Concepts and governing equations for the simulations

The governing flow equations for water flow and nutrient transport are given by the [16] and convection-dispersion equations (CDE) [17-19]:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x_i} \left[K \left(K_{ij}^A \frac{\partial h}{\partial x_j} + K_{iz}^A \right) \right] - s(h) \quad (5)$$

Where θ is the volumetric water content [$\text{L}^3 \text{L}^{-3}$], h is the pressure head [L], x_i ($i = 1, 2$) are the spatial coordinates [L], t is time [T], K_{ij}^A are components of a dimensionless anisotropy tensor K^A (which reduces to the unit matrix when the medium is isotropic), K is the unsaturated hydraulic conductivity function (LT^{-1}), and s is a sink/source term [$\text{L}^3 \text{L}^{-3} \text{T}^{-1}$], accounting for root water uptake (transpiration). The sink/source represents the volume of water removed per unit time from a unit volume of soil due to compensated citrus water uptake.

The equation (CDE) governing transport of independent solutes i.e. single-ion transport is given as:

$$\frac{\partial \rho_b c_1}{\partial t} + \frac{\partial \theta c_2}{\partial t} = \frac{\partial}{\partial x_i} \left(\theta D_{ij} \frac{\partial c_2}{\partial x_j} \right) - \frac{\partial q_i c_2}{\partial x_j} - \Phi - r_a(c, h) \quad (6)$$

Where c_1 and c_2 are solute concentrations in the solid (MM^{-1}) and liquid (ML^{-3}) phases, respectively; q_i is the i^{th} component of volumetric flux density (LT^{-1}), Φ is the rate of change of mass per unit volume by chemical or biological reactions or other sources (negative) or sinks (positive) ($\text{ML}^{-3} \text{T}^{-1}$), respectively, providing connections between individual chain species, ρ_b is the soil bulk density (ML^{-3}), D_{ij} is the dispersion coefficient tensor for the

liquid phase [$\text{L}^2 \text{T}^{-1}$]. The term r_a represents the root nutrient uptake ($\text{ML}^{-3} \text{T}^{-1}$) which is the sum of actual active and passive nutrient uptake. The solid phase concentration, c_1 , accounts for nutrient either sorbed in the solid phase or precipitated in various minerals. This is usually quantified by the adsorption isotherm relating c_1 and c_2 described by the linear equation of the form:

$$c_1 = K_d c_2 \quad (7)$$

Where K_D ($\text{L}^3 \text{M}^{-1}$) is the mass distribution coefficient of species 1. A tracer (e.g. bromide) are assumed to have a $K_D = 0 \text{ cm}^3 \text{ g}^{-1}$. The first order decay constant ranges from 0.36-0.56 d^{-1} [20]. For P, K_D is reportedly in the range of 19 to 185 $\text{cm}^3 \text{ g}^{-1}$ [21,22]. Bulk density for the soil is in the range 1.59-1.72 g cm^{-3} (Immokalee) and 1.55-1.93 g cm^{-3} (Lake Alfred) (T.A. Obreza, unpublished).

The sink term, s , for the Richards equation represents the volume of water removed per unit time from a unit volume of soil due to plant water uptake. Thus, s is defined as:

$$s(h) = \alpha b L_t \frac{T_p}{w} \quad (8)$$

Where the water stress response function $\alpha(h)$ is a prescribed dimensionless function of the soil water pressure head, b is the normalized water uptake distribution, L_t is the width of the soil surface associated with the transpiration process and T_p is the potential transpiration rate (LT^{-1}) and w is the water stress index.

The nonlinear, predictive equations for the unsaturated hydraulic function in terms of soil water retention parameters are given by [23] as:

$$\theta(h) = \begin{cases} \theta_r + \frac{\theta_s - \theta_r}{\left[1 + |\alpha h|^n\right]^m} & h < 0 \\ \theta_s & h \geq 0 \end{cases} \quad (9)$$

$$K(h) = K_s S_e^l \left[1 - (1 - S_e^{1/m})^m\right]^2 \quad (10)$$

Where

$$m = 1 - \frac{1}{n}, \quad n > 1 \quad (11)$$

$$S_e = \frac{(\theta - \theta_r)}{(\theta - \theta_s)} \quad (12)$$

Where θ_r , θ_s , K_s and l are residual water content ($\text{L}^3 \text{L}^{-3}$), saturated water content ($\text{L}^3 \text{L}^{-3}$), saturated hydraulic conductivity (LT^{-1}), and pore connectivity parameter (estimated to be an average of 0.5 for many soils respectively). Parameters α (L^{-1}) and n are empirical coefficients affecting the shape of the hydraulic functions [23]. We estimated the hydraulic functions α and n after fitting the water content and matric potential data using the

van Genuchten model in Community Analyses System (CAS) 2007 [24] developed for determination of soil hydraulic functions (Table 3).

Cumulative flux and concentration

The P concentration and flux were predicted at 30 cm soil depth. Assuming isotropy and a homogeneous profile, the concentration and fluxes at 60 cm soil depth were also predicted.

Results and Discussion

Characterizing the P isotherms

The results of P adsorption were described by a Freundlich

Table 3: Soil water retention parameters of Immokalee and Candler fine sand estimated using CAS software developed by Bloom (2009) [24].

Soil	Depth (cm)	α (cm ⁻¹)	n	m	η
Immokalee	0-15	0.03	1.87	0.47	0.5
Immokalee	15-30	0.04	1.29	0.23	0.5
Immokalee	30-45	0.03	2.06	0.52	0.5
Immokalee	45-60	0.03	1.71	0.42	0.5
Candler	0-15	0.03	2.22	0.55	0.5
Candler	15-30	0.04	1.70	0.41	0.5
Candler	30-45	0.02	2.50	0.60	0.5
Candler	45-60	0.02	1.82	0.45	0.5

η Pore connectivity parameter (estimated to be an average of 0.5 for many soils [18].

lich model with linearized K_D ranging from 0.50 ± 0.19 to 0.75 ± 0.13 kg L⁻¹ for Immokalee fine sand and from

Table 4: Sorption coefficients for P on Immokalee and Candler fine sand using three electrolytes.

Soil	Depth (cm)	Supporting electrolyte	* K_D (L kg ⁻¹)
Immokalee	0-15	0.01 M KCl	0.53 ± 0.11
Immokalee	15-30	0.01 M KCl	0.50 ± 0.19
Candler	0-15	0.01 M KCl	2.87 ± 0.43
Candler	15-30	0.01 M KCl	3.79 ± 0.87
Immokalee	0-15	0.005 M CaCl ₂	0.75 ± 0.13
Immokalee	15-30	0.005 M CaCl ₂	0.74 ± 0.32
Candler	0-15	0.005 M CaCl ₂	3.46 ± 0.65
Candler	15-30	0.005 M CaCl ₂	4.43 ± 0.50
Immokalee	0-15	Fertilizer mixture	0.45 ± 0.10
Immokalee	15-30	Fertilizer mixture	0.43 ± 0.20
Candler	0-15	Fertilizer mixture	1.73 ± 0.15
Candler	15-30	Fertilizer mixture	2.05 ± 0.89
Statistics			
Soil type (S)			***
Electrolyte (E)			***
Depth (D)			***
S × D			***
S × E			***
E × D			***
E × S × D			***

* K_D : Linearized K_D using Equation 12 presented as mean \pm one standard deviation of 3 replications and a C_{max} of 15 mg L⁻¹; ***, 'Significant at 0.001 probability level'.

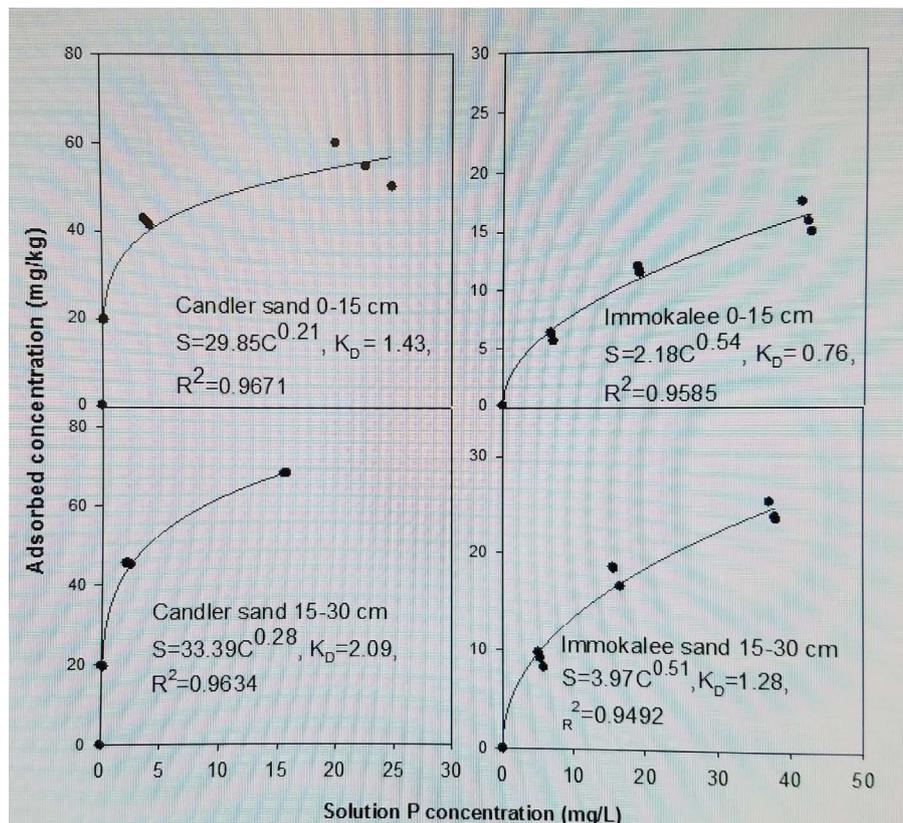


Figure 1: Selected Freundlich isotherms for P for Immokalee and Candler sand using 0.005 M CaCl₂.

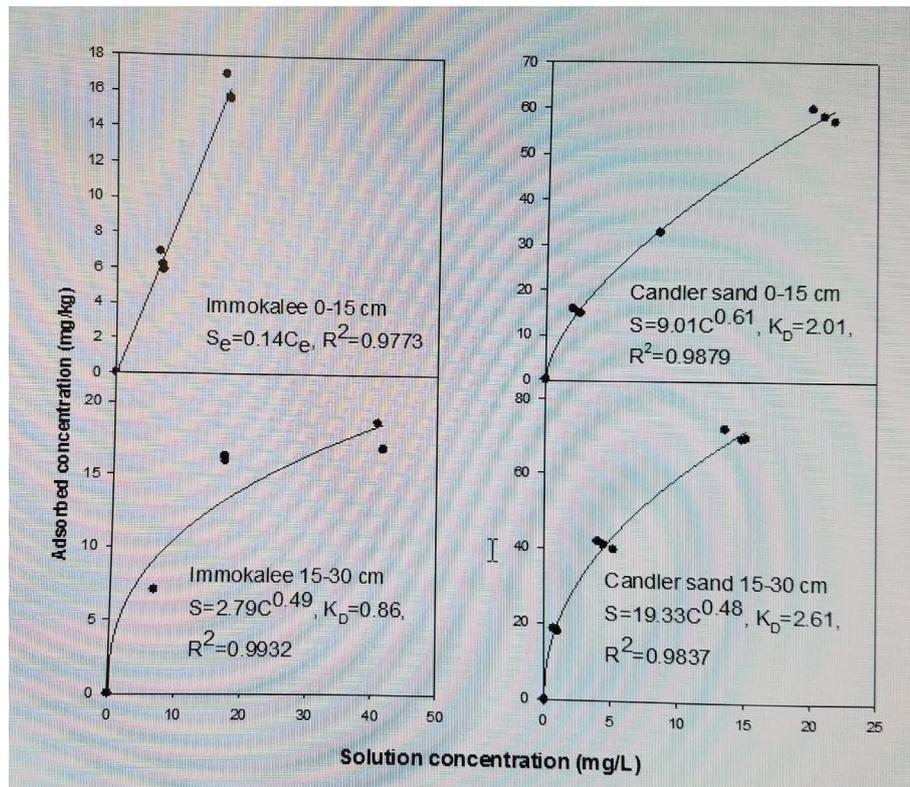


Figure 2: Selected linear and Freundlich isotherms for P for immokalee and candler sand using 0.01 M KCl.

1.73 ± 0.15 to 4.43 ± 0.50 kg L⁻¹ for Candler fine sand using a C_{max} of 15 mg L⁻¹ (Figure 1 and Figure 2). Phosphorus adsorption was linear for the P concentration range (0 to 50 mg P L⁻¹) studied on the Immokalee sand using fertilizer mixture with K_D averaging about 0.44 ± 0.10 kg L⁻¹ (Table 4). Freundlich sorption coefficients (K_f) were lower for Immokalee fine sand than for Candler. High coefficients were observed on Candler fine sand with K_f values eightfold greater than that of Immokalee fine sand. The K_f value obtained with 0.005 M CaCl₂ was approximately twofold that obtained with 0.01 M KCl and threefold that obtained in the fertilizer mixture suggesting the influence of the cation effect on P adsorption than with water. According to [7], the lower Freundlich sorption coefficients (K_f), indicate low P retention capacity at low P concentrations suggesting that the potential risk of subsurface P movement and leaching would be high when the concentration of P in surface soils is high. The K_f and K_D values reported in were generally lower than those reported for carbonatic soils in south Florida [7] where K_D ranged from 14.8 to 76.3 L kg⁻¹ and K_f from 12 to 58 mg^{1-N} kg⁻¹ L^N. However, the results in this study agree with those of other researchers [10,13,25]. According to [13], divalent cations on the CEC enhance P adsorption relative to monovalent cations because they increase the accessibility of (+)-charged edges of clay minerals to P. This occurs at pH < 6.5 (the pH of our soils ranged from 4.9 to 5.6, Table 1, because at greater

soil pH Ca-P minerals would precipitate while at lower pH (< 4.5), Al-P and Fe-P compounds dictate P adsorption. Barrow, et al. (1980) [25] also showed that at equal ionic strength below pH = 6, there was more phosphate adsorption from CaCl₂ than from NaCl on goethite. This phenomenon, according to Barrow and colleagues [25], is caused because high concentration of positive charges near the negatively charged soil surface may be induced by replacing a monovalent cation with a divalent one and also if the added divalent cation has a specific affinity for the adsorption surface. Addition of cations from the supporting electrolyte, unlike using the fertilizer, induced a greater positive charge for phosphate (and orthophosphate anion) adsorption. The higher K_Ds for Candler fine sand might be due to high organic matter and some Fe/Al coatings that might bind P. The high K_f value in the top 0-15 cm than the 15-30 cm layer is ascribed to higher organic carbon and organic matter in the former layer resulting in increased P adsorption. Our results are comparable to those reported on Margate sand and Immokalee fine sand by Muwamba, et al. (2016) [26].

Phosphorus movement with microsprinkler irrigation as function of time

Phosphorus movement was predicted using three different K_Ds estimated with fertilizer mixture, 0.01 M KCl and 0.005 M CaCl₂ for of 21 days, assuming no rainfall events (Figure 3). The assumption is that a K_D value ob-

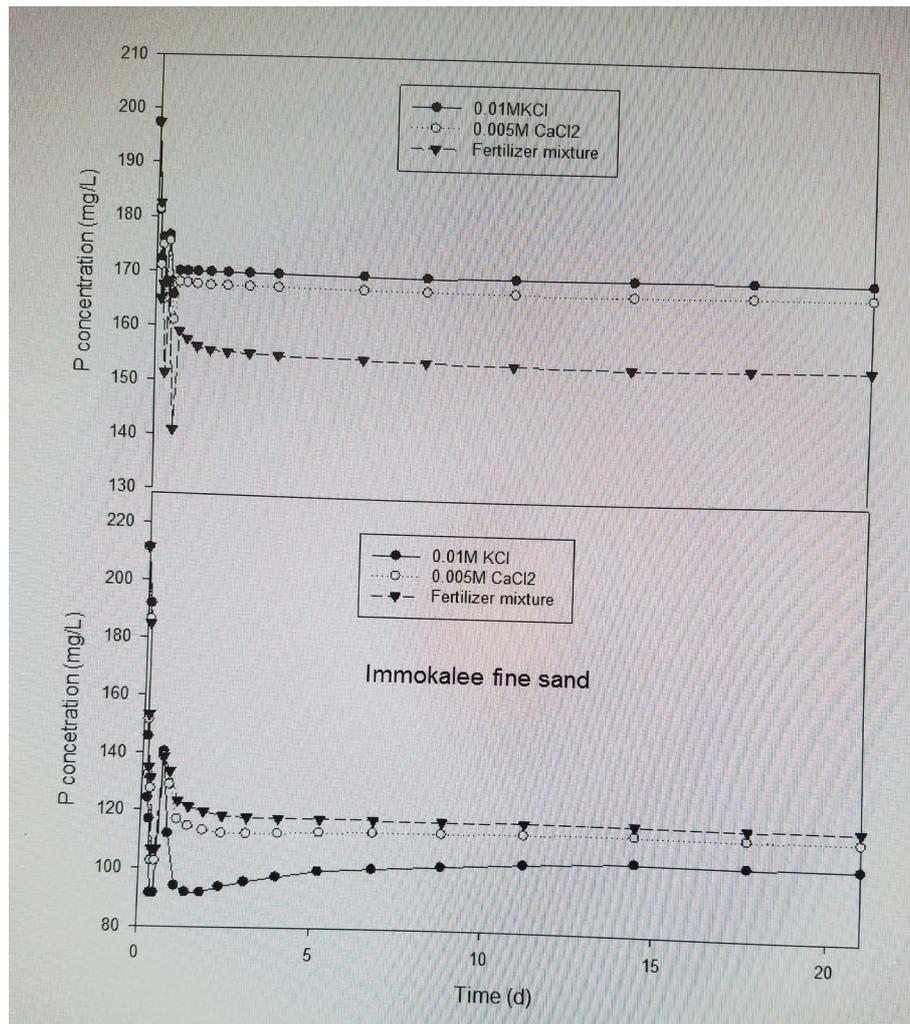


Figure 3: Phosphorus movement on candler and immokalee fine sand depending on the sorption coefficient (K_D) value estimated using HYDRUS-1D.

tained using fertilizer mixture typifies that of field conditions with regard to chemical processes. The results on Candler fine sand at Lake Alfred showed that that P contents for the K_D estimated with 0.01 M KCl and 0.005 M CaCl_2 were 10 to 15% higher than those predicted with a K_D value measured with fertilizer mixture. The predictions on Immokalee fine sand showed that that P contents for the K_D estimated with fertilizer mixture and 0.005 M CaCl_2 were 12 to 20% higher than those predicted with a K_D value measured with 0.01 M KCl. The outputs with K_D measured with 0.005 M CaCl_2 appear to be close to those predicted with a K_D measured with fertilizer mixture on Immokalee fine sand. Thus, the analysis of the K_D values across all electrolytes on the two soils studied revealed that 0.01 M KCl is the electrolyte that yields K_D values (0.42 to 0.69 L kg^{-1} on Immokalee fine sand and 0.42 to 1.06 L kg^{-1} on Candler fine sand) fairly close to fertilizer mixture on coated sands while 0.005 M CaCl_2 tends to give K_D values (2.44 to 4.66 L kg^{-1} on Immokalee fine sand and 2.81 to 4.93 L kg^{-1} on Candler fine

sand) two to threefold in magnitude to those determined with fertilizer mixture on sands with Fe coatings. This suggests that 0.005 M CaCl_2 would tend to overestimate P sorption and retardation during unsaturated or saturated flow on coated sand while estimating the adsorption process satisfactorily on uncoated sands than 0.01 M KCl. It appears the addition of a supporting electrolyte with a divalent or monovalent cation, unlike fertilizer mixture, increases the surface charge for adsorption of orthophosphate anions on sands with coatings. The use of 0.01 M KCl appears not to be influenced by presence of sand coatings compared with CaCl_2 and, thus, would present an appropriate supporting electrolyte for Candler fine sand while CaCl_2 would be appropriate for the Immokalee fine sand.

Conclusion

The results show that P adsorption in the top 0-15 cm was greater for Candler than Immokalee sand using the fertilizer mixture, 0.005 M CaCl_2 and 0.01 M KCl. The

mass distribution coefficients (K_D) for P estimated using 0.01 M KCl were similar to values determined using fertilizer mixture for Candler fine sand. The K_D values determined using 0.005 M CaCl_2 as the supporting electrolyte were two- to threefold greater than the K_D of the fertilizer mixture on Candler fine sand suggesting that divalent Ca might result in overestimation of P sorption on Candler sandy soils. On Immokalee fine sand, fertilizer mixture and 0.005 M CaCl_2 were comparable and resulted in greater P adsorption than 0.01 M KCl. Thus, it would be appropriate to use 0.01 M KCl as supporting electrolyte for Florida's Candler fine sand and 0.005 M CaCl_2 on Immokalee fine sand.

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